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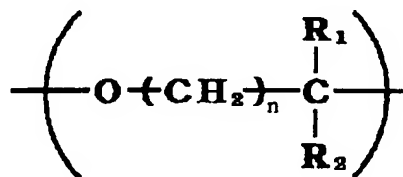
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(54) [Title of invention]

Resin composition

(57) [Summary]

[Method for solution] Resin composition which contains aliphatic group polyester for (A), polyalkylene glycol for component (B) shown by formula (1) (in the formula (1), R<sub>1</sub> and R<sub>2</sub> are independent, saturated or unsaturated hydrocarbon groups of which hydrogen atom or carbon atom are 1 to 10 and n shows integer of 0 to 5), aliphatic carboxylic acid amide for component (C) and inorganic additive containing SiO<sub>2</sub> having average particle diameter of less than 30 μm for component (D).

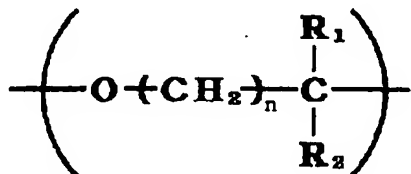


[Effect] The resin composition concerning this invention is superior in forming ability in forming (for example, such as extrusion forming and inflation forming) when compared with the case made with single component (A). Also the film and sheet concerning this invention is significantly superior in anti blocking and slippage compared with the case made by single component (A).

**[Claim]**

**[Claim 1]** Resin composition which contains aliphatic group polyester for (A), polyalkylene glycol for component (B) shown by formula (1) ((chemical formula 1) in the formula (1),  $R_1$  and  $R_2$  show independent saturated or unsaturated hydrocarbon groups of which hydrogen atom or carbon atom are 1 to 10 and  $n$  shows integer of 0 to 5), aliphatic carboxylic acid amide for component (C) and inorganic additive containing  $SiO_2$  having average particle diameter of less than 30  $\mu m$  for component (D).

[Chemical formula 1]



**[Claim 2]** Resin composition described in Claim 1 of which aliphatic group polyester of component (A) is poly lactic acid.

**[Claim 3]** Resin composition described in Claim 1 which is characterized by at least one kind of aliphatic group polyester of component (A) is selected from a group of (a1) hydroxy carboxylic acid (a2) polyhydroxy carboxylic acid and (a3) aliphatic group polyhydric alcohol and polybasic acid and (a4) aliphatic group polyester of aliphatic group polyhydric alcohol and aliphatic group polybasic acid, and manufactured by dehydration (polymerized) condensation reaction.

**[Claim 4]** Resin compound described in Claim 1 which is characterized by at least one kind of aliphatic group polyester of component (A) is selected from a group of (a1-1) lactic acid, (a1-2) glycolic acid, (a2-1) poly lactic acid, (a2-2) polyglycolic acid, (a3-1) ethylene glycol and succinic acid, (a3-2) ethylene glycol and adipic acid, (a3-3) ethylene glycol, succinic acid and adipic acid, (a3-4) ethylene glycol, 1,4-butane diol

and succinic acid, (a3-5) ethylene glycol, 1,4-butane diol and adipic acid, (a3-6) ethylene glycol, 1,4-butane diol, succinic acid and adipic acid, (a3-7) 1,4-butane diol and succinic acid, (a3-8) 1,4-butane diol and adipic acid, (a3-9) 1,4-butane diol, succinic acid and adipic acid, (a4-1) polyethylene succinate and (a4-2) polybutylene succinate and manufactured by dehydrating (poly) condensation reaction.

**[Claim 5]** Resin compound described in Claim 4 which is characterized by that weight of (a1-1) lactic acid and/or (a2-1) poly lactic acid is more than 50 weight % to component (A) aliphatic group polyester.

**[Claim 6]** Resin compound described in Claim 1 which is characterized by that aliphatic group polyester of component (A) is a mixture of more than 50 weight % poly lactic acid and less than 50 weight % of aliphatic group polyesters other than poly lactic acid.

**[Claim 7]** Resin composition described in any of Claim 1 through 6 characterized by that polyalkylene glycol of which component (B) is shown by the formula (1) chemical formula (1) is polyethylene glycol and/or polyethylene-polypropylene glycol.

**[Claim 8]** Resin composition described in any of Claim 1 through 7 characterized by that aliphatic group carboxylic acid amide of component (C) is at least one kind selected from a group of stearic acid amide, oleic acid amide, behenic acid amide, erucic acid amide, N-oleyl palmito amide and N-stearyl erucic acid amide.

**[Claim 9]** Resin composition described in any of Claim 1 through 8 characterized by that amount of polyalkylene glycol shown by (chemical formula 1) of the formula (1) of component (B) to the weight of resin composition is 3 to 50 weight %, aliphatic group carboxylic amide of component (C) is less than 0.01 to 10 weight % to the weight of resin composition and inorganic additive of component (D) containing  $SiO_2$  having

less than 30  $\mu\text{m}$  average particle diameter is 0.01 to 10 weight % to the weight of resin composition.

**[Claim 10]** Formed product made of resin composition described in any of Claim 1 through Claim 9.

**[Claim 11]** Film made by forming resin composition described in any of Claim 1 through Claim 9.

**[Claim 12]** Sheet made by forming resin composition described in any of Claim 1 through Claim 9.

**[Detailed explanation of the invention]**

**[0001]** [Technical field where this invention belongs] This invention relates to the resin composition having flexibility and sometimes transparency, excellent anti blocking property and opening property at the time of extrusion forming and inflation forming,, and being biodegradable in natural environment. Furthermore, this invention relates to the formed body such as film, sheet and bag for packaging obtained from forming.

**[0002]**

[Conventional technology] Plastics increases the volume of garbage when discarded after use and furthermore, it has a problem of remaining in the soil semi-permanently because it is almost not biodegradable even buried in natural environment. It has been the environmental issue by destroying the natural view and living environment of marine lives . Some measures of recycling the plastics were considered against these problems such as a collecting system by classifying the disposals, returning system and deposit system, however the realization of these measures is not penetrated to the end user at this moment.

**[0003]** On the other hand, biodegradable plastics which has a biodegrading function by microorganisms existent in natural environment has caught attention and noticed as being capable for solving above mentioned environmental issues. Already, many environmentally effective products

which used this biodegradable plastics are suggested and for example, it is applied in packaging container field, hard container for drink and shampoo and soft packaging for snacks and furthermore, container for liquid by a complex material which is combined with paper. In the other field, it is introduced to plastic dish, stationery and sundries and there are many already commercially available. Some local government are trying the plastic bag for collecting kitchen scraps which is made of biodegradable plastics.

**[0004]** For biodegradable plastics having biodegradable function, there are one kind which is a mixture with other plastics with natural substances of polysaccharide and sugar such as cellulose and starch, one kind which is synthesized by microorganism and another kind which is chemically synthesized. Other biodegradable plastics which used (poly)saccharide of starch, chitin or bacteria cellulose has aliphatic polyester as a main body. For aliphatic group polyester produced by microorganism, the one having hydroxyalcanoate is known and 3-hydroxybutyrate-3-hydroxyvalerate are sold as brand name [Biopol] by Monsanto. For chemically synthesized one, there are polycaprolacton, polybutylene succinate and poly lactic acid which was made of lactic acid as a main component.

**[0005]** For biodegradable plastics which can be used as a material of molded product such as film, sheet and packaging bag which are the objects of this invention, it is necessary to be flexible, superior in transparent property in some case and excellent in workability of melt cut and deposition. However, in aliphatic group polyester, there are some which are hard and lacking flexibility and for the purpose of using for film sheet and packaging bag of this invention, flexibility needs to be added. Generally, for the method of adding flexibility, a method to add plasticizer, a method to make it soft by copolymerization and another method to

mix other soft polymer are known.

[0006] The method of adding plasticizer or softening by copolymerization satisfies above mentioned objective, however, the former method may have problems such as variation over the time and deterioration of secondary workability by the bleeding of plasticizer and the latter method has a problem of complicated manufacturing process. In comparison to this, the other method to mix other soft polymer does not cause a change over the time and can be employed as an easy and reasonable method.

[0007] For soft type polymers which satisfies above mentioned requirement in this method, polyalkylene glycol such as polyethylene glycol (abbreviated as PEG, hereinafter) and polypropylene glycol are listed. It is already disclosed that these polymers can add flexibility without damaging transparency which is an excellent property of poly lactic acid. (European Patent Publication 0515203)

[0008]

[Problems to be solved by the invention] However, when the mixture of aliphatic polyester (poly lactic acid, for example) and polyalkylene glycol (polyethylene glycol (PEG)) is formed into film or sheet shape by T die extrusion forming and inflation forming, even the obtained film has superior flexibility, there are practical problems of: locating to the screw heads is unstable and forming ability is bad, obtained rolled film causes blocking and bag can not be opened in case of inflation film. Furthermore, when PEG having high molecular weight (1,000,000 to 5,000,000) is used, there is additional problem which is reduction of quality in transparency in addition to the above mentioned problems.

[0009] As explained, although the method to add PEG to aliphatic polyester can add flexibility, it is not satisfactory industrially in practical aspect of forming ability into film and sheet. Also, depending on the

amount of molecule of PEG, it is not satisfactory in the area of transparency. Therefore, how to improve the mixture of aliphatic group polyester and alkylene glycol to composition having anti blocking property and slippage (opening property) was the objective.

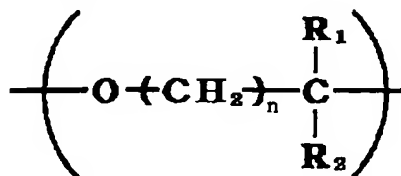
[0010]

[Method to solve the subject] After considering above mentioned problems and serious investigation, inventors discovered that by adding a specific inorganic additive and aliphatic group carboxylic acid amide to the mixture of aliphatic group polyester and alkylene glycol, above mentioned problems can be solved and was completed this invention.

[0011] Following is the detailed explanation of this invention. Aliphatic group polyester in this invention means homopolymer, copolymer and the mixture of aliphatic group polyester made of hydroxy carboxylic acid or aliphatic group polyhydric alcohol and aliphatic group polyhydric carboxylic acid. These polymers could be the one of which chain is extended partially by binder such as isocyanate and crosslinking agent such as polysaccharide. This invention is specified by the followings.

[1] Polyalkylene glycol shown by aliphatic group polyester for component (A) and component (B) of formula (1)(chemical formula 2).

[0012] [Chemical formula 2]



(in the formula (1)(chemical formula 2), R<sub>1</sub> and R<sub>2</sub> are independent, saturated or unsaturated hydrocarbon groups of which hydrogen atom or carbon atom are 1 to 10 and n shows integer of 0 to 5), aliphatic group carboxylic acid amide for component (C)

and inorganic additive containing  $\text{SiO}_2$  and having average particle diameter of less than  $30\ \mu\text{m}$  for component (D).

**[0013]** [2] Resin composition described in [1] of which aliphatic group polyester of component (A) is poly lactic acid.

[3] Resin composition described in Claim 1 which is characterized by at least one kind of aliphatic group polyester of component (A) is selected from a group of (a1) hydroxy carboxylic acid (a2) polyhydroxy carboxylic acid and (a3) aliphatic group polyhydric alcohol and polybasic acid and (a4) aliphatic group polyester of aliphatic group polyhydric alcohol and aliphatic group polybasic acid, and manufactured by dehydration (polymerized) condensation reaction.

**[0014]** [4] Resin compound described in Claim 1 which is characterized by at least one kind of aliphatic group polyester of component (A) is selected from a group of (a1-1) lactic acid, (a1-2) glycolic acid, (a2-1) poly lactic acid, (a2-2) polyglycolic acid, (a3-1) ethylene glycol and succinic acid, (a3-2) ethylene glycol and adipic acid, (a3-3) ethylene glycol, succinic acid and adipic acid, (a3-4) ethylene glycol, 1,4-butane diol and succinic acid, (a3-5) ethylene glycol, 1,4-butane diol and adipic acid, (a3-6) ethylene glycol, 1,4-butane diol, succinic acid and adipic acid, (a3-7) 1,4-butane diol and succinic acid, (a3-8) 1,4-butane diol and adipic acid, (a3-9) 1,4-butane diol, succinic acid and adipic acid, (a4-1) polyethylene succinate and (a4-2) polybutylene succinate and manufactured by dehydrating (poly) condensation reaction.

**[0015]** [5] Resin compound described in [4] which is characterized by that weight of lactic acid (a1-1) and/or poly lactic acid (a2-1) is more than 50 weight % to component (A) aliphatic group polyester.

[6] Resin compound described in [1] which is characterized by that aliphatic group polyester of component (A) is a mixture of more than 50 weight % poly lactic acid and

less than 50 weight % of aliphatic group polyesters other than poly lactic acid.

**[0016]** [7] Resin composition described in any of [1] through [6] characterized by that polyalkylene glycol of which component (B) is shown by the formula (1), chemical formula (2) is polyethylene glycol and/or polyethylene-polypropylene glycol.

[8] Resin composition described in any of [1] through [7] characterized by aliphatic group carboxylic acid amide of component (C) is at least one kind selected from a group of stearic acid amide, oleic acid amide, behenic acid amide, erucic acid amide, N-oleyl palmito amide and N-stearyl erucic acid amide.

**[0017]** [9] Resin composition described in any of [1] through [8] characterized by that amount of polyalkylene glycol shown by (chemical formula 2) of the formula (1) of component (B) to the weight of resin composition is 3 to 50 weight %, aliphatic group carboxylic amide of component (C) is less than 0.01 to 10 weight % to the weight of resin composition and inorganic additive of component (D) containing  $\text{SiO}_2$  having less than  $30\ \mu\text{m}$  average particle diameter is 0.01 to 10 weight % to the weight of resin composition.

**[0018]** [10] Formed product made of resin composition described in any of [1] through [9].

[11] Film made by forming resin composition described in any of [1] through [9].

[12] Sheet made by forming resin composition described in any of [1] through [9].

**[0019]** [Aliphatic group hydroxy carboxylic acid] For the concrete example of aliphatic hydroxy carboxylic acid used in this invention, glycolic acid, lactic acid, 3-hydroxybutyric acid, 4-hydroxyvaleric acid, 5-hydroxyvaleric acid and 6-hydroxycaproic acid are listed for example.

**[0020]** [Aliphatic group polyhydric caproic acid and its anhydride] Aliphatic group polyhydric alcohol and aliphatic group

polyester made of aliphatic group polyhydric carboxylic acid which can be used for the method of this invention can be manufactured by dehydrating and (poly) condensing one or more than two kinds of aliphatic group polyhydric carboxylic acid (preferably aliphatic group dicarboxylic acid) and/or their anhydride and one or more than two kinds of aliphatic group polyhydric alcohol (preferably, aliphatic group diol).

[0021] For aliphatic group polyhydric carboxylic acid and its anhydride in this case, oxalic acid, succinic acid, malonic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azaleic acid, sebacic acid, aliphatic group dicarboxylic acid and its anhydride of such as undecane dioxide and dodecane dioxide are listed.

[0022] [Aliphatic group polyhydric alcohol] For the concrete examples of aliphatic group polyhydric alcohol, there are such as ethylene glycol, diethylene glycol, triethylene glycol, propylene glycol, dipropylene glycol, 1,3-butane diol, 1,4-butane diol, 3-methyl-1,5-pentanediol, 1,6-hexanediol, 1,9-nonane diol, neopentyl glycol, tetramethylene glycol and 1,4-cyclohexane dimethanol.

[0023] [Polysaccharide] For the crosslinking agent of aliphatic group polyester, polysaccharide having multifunctional radical is listed. For the concrete example of polysaccharide, cellulose, cellulose nitrate, cellulose acetate, methyl cellulose, ethyl cellulose, celluloid, viscose rayon, regenerated cellulose, cellophane, cupra, cuprammonium rayon, cuprophane, Bemberg, hemicellulose, starch, amylopectin, dextrin, dextran, glycogen, pectin, chitin, chitosan, gum arabic, guar gum, locust bean gum, acacia gum and their mixture and their derivatives are listed, especially, acetyl cellulose and ethyl cellulose are preferably used.

[0024] [Kind of aliphatic group polyester] Aliphatic group polyester shown in this invention may be homopolymer, random

copolymer and block copolymer and the mixture of more than two kinds of their homopolymer which are obtained by dehydrating and condensing with above mentioned hydroxycarboxylic acid and/or aliphatic group polyhydric carboxylic acid and aliphatic group polyhydric alcohol.

[0025] The mixture of copolymer consisted of lactic acid of which poly lactic acid and lactic acid content rate is more than 50 weight % and other hydroxy carboxylic acid or aliphatic polyhydric alcohol and aliphatic polyhydric carboxylic acid and poly lactic acid of which lactic acid content is more than 50 weight % and aliphatic group polyester other than poly lactic acid, is preferable, furthermore, the copolymer consisted of lactic acid of which poly lactic acid and lactic acid content is more than 50 % and other hydroxy carboxylic acid or aliphatic group polyhydric alcohol and aliphatic group polyhydric carboxylic acid is especially preferable because it is excellent in transparency.

[0026] [Manufacturing method of aliphatic group polyester] Manufacturing method of aliphatic group polyester of this invention is not limited especially. For concrete example of manufacturing method, it can be a direct dehydrating condensation method which dehydrate and condensate lactic acid or hydroxy carboxylic acid other than lactic acid or aliphatic group diol and aliphatic group dicarboxylic acid under the existence of organic solvent and catalyst, which was referred by patent publication Toku Kai Shou 6-6536 as shown in an example of synthesis which will be mentioned later. For example, by referring to the publication of US patent 2,703,316, a method of indirect polymerization method may be used wherein after lactic acid or hydroxy carboxylic acid other than lactic acid and lactic acid are dehydrated at first and made into cyclic dimer and ring-opening polymerized. In case of copolymer, random polymer and

block polymer show different mechanical property by such as mixing ratio of each polymer and molecular weight at the time of mixing, however, in this invention, there is not limitation in the mixing ratio and molecular weight and an appropriate polymer composition which is suitable for the targeted use, can be selected.

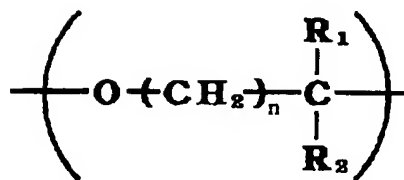
**[0027]** [Molecular weight of aliphatic group polyester] Molecular weight of aliphatic group polyester of this invention is not especially limited if it shows substantially sufficient mechanical property. Generally as weight average molecular weight, 10,000 to 5,000,000 and 30,000 to 3,000,000 are preferable, 50,000 to 2,000,000 is more preferable and 70,000 to 1,000,000 is even more preferable and 90,000 to 500,000 is the most preferable. If a weight average molecular weight is smaller than 10,000, mechanical property may not be sufficient and on the contrary, if a molecular weight is greater than 5,000,000, handling may be difficult or it may not be economical.

**[0028]** Also, weight average molecular weight and molecular weight distribution of aliphatic group polyester can be controlled in a desired range by properly choosing reacting condition such as kind of solvent, kind and amount of catalyst, reaction temperature, reaction time and degree of dehydration of the reaction group.

**[0029]** [Polyalkylene glycol] In this invention, polyalkylene glycol is added in order to add flexibility to aliphatic group polyester. Polyalkylene glycol shown in this invention is a polymer which is shown by formula (1), (chemical formula 3), polyethylene glycol which is solid form in normal temperature and polyethylene polypropylene glycol can be used conveniently as they are reasonable and easily accessible.

**[0030]**

[Chemical formula 3]



(In the (formula 1) and (chemical formula 3), R<sub>1</sub> and R<sub>2</sub> are independent, saturated or unsaturated hydrocarbon groups of which hydrogen atom or carbon atom are 1 to 10 and n shows integer of 0 to 5)

[Molecular weight of polyalkylene glycol] Weight average molecular weight of polyalkylene glycol is preferably 1,000 to 1,000,000 and 2,000 to 500,000 is more preferable, 3,000 to 300,000 is even more preferable and 4,000 to 100,000 is the most preferable. Generally, if it is smaller than 1000, it becomes liquid form at normal temperature and bleeding tends to be caused when mixed to aliphatic group polyester and it is not desirable. On the contrary, if it is greater than 1,000,000, compatibility with aliphatic group polyester tends to become bad and when aliphatic group polyester is superior in transparency, the transparency may sometimes be damaged.

**[0031]** [Amount of addition of polyalkylene glycol] Amount of polyalkylene glycol to be added is normally 3 to 50 weight % preferably 5 to 40 weight %, more preferably 10 to 30 weight % to the amount of polymer composing component (components which are added with aliphatic group polyester of component (A) and polyalkylene glycol shown by (formula 1, chemical formula 3) of component (B) is called as [polymer composing component, hereinafter). Generally, if it is less than 3 weight %, effect of adding flexibility is small, on the contrary, if it is greater than 50 weight %, forming ability may become inferior or compatibility of aliphatic group polyester may become bad. In this invention, when obtaining such as film and sheet by normal inflation forming and T die forming with above mentioned

polymer composing component, it is necessary to add inorganic additive and aliphatic group carboxylic amide to add anti-blocking property and slippage.

**[0032]** [Inorganic additive] Inorganic filler shown in this invention is preferably inorganic additive of which particle diameter of  $\text{SiO}_2$  which is its main ingredient, is less than 30  $\mu\text{m}$ . For a concrete example of inorganic additive, such as talc, magnesium silicate,  $\text{SiO}_2$ , kaolinite are listed and especially  $\text{SiO}_2$  is preferably used. These can be mixture of one or more than two kinds and there is no limitation.

**[0033]** Also, if transparency is highly required in the film or sheet, particle diameter of inorganic additive is preferably smaller than 100 nm and even when mixed to polymer composing component which has excellent transparency, its transparency may be virtually maintained. For the concrete example of such inorganic additive, there is such as synthetic  $\text{SiO}_2$  having 1 to 50 nm particle diameter.

**[0034]** [Amount of inorganic additive to be added] The amount of inorganic additive to be added is generally 0.05 to 15 weight % to 100 weight parts polymer composing component and preferably it is 0.5 to 10 weight % and even more preferably 1 to 5 weight %. Optimum amount of addition is selected so that moldability at the time of T die extrusion and inflation forming, anti-blocking property of obtained film and slippage of the film become excellent.

**[0035]** [Aliphatic group carboxylic amide] Aliphatic group carboxylic amide contains [aliphatic acid amide] described in the right column on page 389 to left column on page 391 of [chemical product 10889] (Year 1989, Kagaku Kogyo Nippo Co., Hamacho Nihombashi, Chuo ku, Tokyo). All the description is made to be a portion of disclosure of the Specification of this Patent by clearly identifying the reference and the range of reference and the directory which

can be led directly and primarily by the people of industry by referring to the range of reference which was clarified, all the item and disclosure listed in the Patent Specification.

**[0036]** For the concrete example of aliphatic group carboxylic acid amide, oleic acid amide, stearic acid amide, erucic acid amide, behenic acid amide, N-oleyl palmito amide, N-stearyl erucic acid amide, N,N'-ethylene bis (stearoamide), N,N'-methylene bis (stearoamide) methylol stearoamide, ethylene bis oleic acid amide, ethylene bis behenic acid amide, ethylene bis stearic acid amide, ethylene bis lauric acid amide, hexamethylene bis oleic acid amide, hexamethylene bis stearic acid amide, butylenes bis stearic acid amide, N,N'-dioleoyl sebacic acid, N, N'-dioleoyl adipic acid amide, N, N'-distearyl adipic acid amide, N, N'-distearyl sebacic acid amide, m-xylilene\* (xylene) bis stearic acid amide, N,N'-distearyl isophthalic acid amide, N,N'-distearyl terephthalic acid amide, N-oleoyl oleic acid amide, N-stearyl oleic acid amide, N-stearyl erucic acid amide, N-oleoyl stearic acid amide, N-stearyl stearic acid amide, N-butyl-N' stearyl urea, N-propyl-N' stearyl urea, N-allyl-N' stearyl urea, N-phenyl-N' stearyl urea, N-stearyl-N' stearyl urea, dimethylol\* (dimethylol) oil amide, dimethyl lauric acid amide, dimethyl stearic acid amide are listed. Especially, oleic acid amide, stearic acid amide, erucic acid amide, behenic acid amide, N-oleoyl palmitoamide, N-stearyl erucic acid amide are preferably used. These can be mixture of one kind or more than two kinds.

**[0037]** [Amount of aliphatic group carboxylic acid amide to be added] Amount of aliphatic group carboxylic acid amide to be added is 0.05 to 10 weight parts to 100 weight parts polymer composing component made of aliphatic group polyester and polyalkylene glycol, preferably, it is 0.1 to 7.0 weight parts more preferably. It is 0.3 to 5.00 weight parts, the most preferably 0.5 to

3 weight parts. The optimum amount added is selected appropriately so that the forming ability of T die injection forming and inflation forming which are the goal of the invention, and anti blocking of the obtained film and slippage are the most superior, as is the case of inorganic additive.

**[0038]** [The third component] In this invention, in order to improve various properties of resin composition (such as tension strength, heat resisting property, weather resistance, for example) some additives (such as antioxidant, thermal stabilizer, ultra violet light stabilizer, filler, antistatic agent, surface wetting improving agent, incineration aiding agent and pigment) and a little amount of other resin may be added. By selecting properly the conditions of extrusion and extension according to the purpose, film or sheet made of resin composition of this invention having desirable physical property, optical property, transmission wavelength spectrum, light shielding property and oil resistance can be manufactured.

**[0039]** [Manufacturing method of resin composition] Manufacturing method of polymer composing component is not especially limited and publicly known method used normally for thermoplastic resin can be adopted. After mixing evenly inorganic additive and aliphatic group carboxylic amide and the third component as needed by using such as high speed agitator or low speed agitator, aliphatic group polyester, polyalkylene glycol, a method to melt and mix by uniaxial or multiaxial extruder which have ample mixing capability, can be adopted.

**[0040]** [Manufacturing and forming of film and sheet] Resin composition of this invention is suitable material for the manufacturing of film and sheet. For the method of forming film, sheet plate shape, thermal forming such as inflation forming and T die forming are listed however, there is not limitation in the method. The film and sheet

of this invention contain film and sheet obtained by publicly known forming method and there is no limitation in its shape, size, thickness and design.

**[0041]** (1) Manufacturing technology  
The film and sheet made of the resin composition of this invention can be manufactured by the publicly known and employed method such as extruding method, c-extruding method, calendar method, hot press method, solvent casting method, inflation method, balloon method and tenter method.

**[0042]** (2) Process specification  
In manufacturing process, conditions of such as uniaxial extension magnification, biaxial extension magnification, number of steps of extension, temperature of heat treatment, variable speed of heat treatment temperature, number of cooling roller, positioning style of cooling roller, winding type to cooling roller, cooling roller temperature, and mirror finish of cooling roller surface can be properly specified according to the purpose.

**[0043]** (3) Methodology of quality control  
By adopting publicly known and employed instrumentation engineering method which adopted such as radiation, electromagnetic wave, light and ultra sound, the data of product thickness is detected and the data is fed back to the manufacturing process and variation in thickness of the product is quality controlled manually or automatically controlled. For the method of instrumentation engineering, alpha beam thickness measuring device, of transmission type (absorbing type) or diffusion type, beta beam thickness measuring device, gamma beam thickness measuring device are included and publicly known and employed radiation isotope is used for the source of beam.

**[0044]** (4) Methodology of post treatment process and finishing process  
In post treatment process or finishing

process, post treatment processes and finishing process of such as welding, heat seal, perforating, primer coating, adhesive coating chemical coating, Parkalizing, vapor deposition, sputtering, (CVD) coating, etching, spraying, dying, painting, static coating, air brushing, laminate, sandwich, embossing addition, three dimensional pattern adding, embossing, waving, printing, transferring, sanding, sandblast, shearing, punching, blanking, honeycomb structuring, corrugated structuring and layer forming can be done. In post treatment process or finishing process, publicly known or employed methods such as calendar method, extrusion method, screen printing method, gravure printing method, relief printing, intaglio printing, doctor blade method, soaking method, spray method, air brush method and electrostatic coating method can be adopted. The film or sheet made of the resin composition of this invention can be multi-layer structured layer by laminating or bonding with the sheet of other material such as paper and other polymer.

**[0045]** (5) Methodology of extrusion method or co-extrusion method

In extrusion method or co-extrusion method, publicly known and employed die such as T die, inflation die (circular die), flat die, feed block/single manifold die and single manifold die combined with several feed blocks can be used. In co-extruding method, multiple polymer of different character and or other kind of polymer are used and multi layered film can be manufactured.

**[0046]** When inflation method or balloon method is used, as biaxial simultaneous extension is enabled, sturdy product having low extension rate, high elasticity and sturdiness can be manufactured at high production rate and relatively reasonable cost and also, as it is a bag shape (seamless form) it is appropriate for the manufacturing of pouch and bag shape packing bag for super market, bags preventing wetting from

the condensed water on the package of such as frozen food and meat at low temperature and compost bag. By combining with co-extrusion method, multiple layer film can be manufactured at high productivity by using multiple resin composition of this invention having different characteristics and/or other kind of polymer. Inflation method or balloon method can be combined with co-extrusion method. Film or sheet made of resin composition of this invention can be manufactured into roll form, tape form, cut sheet form, plate form and bag form (seamless form) by setting the condition of process according to the purpose.

**[0047]** (6) Secondary process

The film or sheet made of resin composition of this invention is also appropriate for the secondary process which generates two dimensional or three dimensional form such as extension processing, blow processing and vacuum forming.

**[0048]** (7) Concrete example of use

The film or sheet made of the resin composition of this invention can be conveniently used as shopping bag, garbage bag, compost bag, food and snack wrapping film, food wrapping film, wrapping film for cosmetics and fragrance, wrapping film for medical use, wrapping film for crude drug, medical wrapping film for surgical use for stiff shoulder and sprain, agricultural and gardening film, wrapping film for agricultural chemical, film for greenhouse, bag for fertilizer, wrapping film for magnetic tape and cassette product such as video and audio tape, wrapping film for floppy disc, film for overhead projector, film for plate making, tracing film, fence, oil fence for marine, river, lake and pond, adhesive tape, tape, bonding material water proof sheet, umbrella, tent, bag for soil and bag for cement.

**[0049]** Film or sheet which is one style of the formed body of this invention can be conveniently used by using its property especially in the use which requires trans-

parency and degrading. When the film or sheet which is one style of the formed body of this invention is used as wrapping material for the bag to put food and snack, term of storage and shelf life can be extended greatly by including oxygen absorbing agent when sealing the food and snacks in the bag.

**[0050]** Following is the embodiment examples, however, unless exceeding beyond the technical range of this invention it is not limited to those. Also, weight average molecular weight (Mw) in the embodiment examples is measured by comparing to the polystyrene standard sample by gel permeation chromatography (column temperature 40°C, chloroform solvent). Also, physical properties on the obtained film is measured with the following condition.

(1) Modulus of elasticity: Followed JIS-K-6732

(2) Anti blocking property: Followed JIS-Z-0219

(3) Slippage: By using a friction measuring device made by Toyo Seiki, the sample is applied to the tilted plate and block (1 kg load) and the plate is tilted at the constant speed (2.7 °/sec) from the level position and the angle in which block starts slipping was measured.

**[0051]** [Example of synthesis 1] In four reacting ovens equipped with agitator and thermometer, 104.3 kg of 90% -lactic acid and 225.0 kg diphenyl ether and 2.0 kg metallic tin are put and heated and stirred while generated water was distilled outside the system at 130 °C/140mmHg for 7 hours. Dean Stark Trap was installed to this and azeotrope dehydrated for 8 hours at 140 °C/130mmHg for 8 hours then, drying tube filled with 40kg molecular sieves 3A was installed and heated and refluxed at 130 °C/17mmHg for 30 hours so that distilled solvent would return to the reaction device through the drying tube. Reacted mass was dissolved into 600 litre chloroform after

cooling and added to 4k litre acetone and precipitated again, then deposited solid was filtered. Then the solid was added with isopropyl alcohol solution dissolved with 5 kg hydrochloric acid (abbreviated as IPA, hereafter) and stirred for 30 minutes and furthermore, 500 litre IPA was added and filtered after sludging, this was repeated for 3 times and obtained wet cake was dried at 60 °C/100mmHg for 15 hours. Obtained solid was white powder form polylactic acid and the obtained amount was 69.1 kg and the yield was 92.2 % and weight average molecular weight (Mw) was 155,000.

**[0052]** [Example of synthesis 2] Similar method was adopted except using 50.5 kg 1,4-butane diol and 66.5 kg succinic acid, instead of lactic acid and as a result, white powder form polybutylene succinate was obtained. The obtained amount was 92.2 kg and the yield was 95.0 % and weight average molecular weight (Mw) was 122,000.

**[0053]** [Example of synthesis 3] 8 kg polylactic acid obtained from the synthesis example 1 and 2 kg polybutylene succinate obtained from synthesis example 2 were mixed well and a mixture of polylactic acid and polybutylene succinate was obtained.

**[0054]** [Example of synthesis 4] Other than that 9.05 kg polybutylene succinate having 120,000 weight average molecular weight (Mw) obtained from synthesis example 2 was added to the reaction mass of polylactic acid having 22,000 weight average molecular weight obtained by the same method with synthesis example 1 (polylactic acid for 75 kg and diphenyl ether for 225.50 kg) and furthermore, reacted for 20 hours at 130 °C/17mmHg, the rest was followed similar to the synthesis example 1 and block copolymer of polylactic acid and polybutylene succinate (content of lactic acid was 89 %) was obtained as a result. The amount obtained was 78.3 kg, yield was 93.2 % and weight average molecular weight (Mw) was 135,000.

[0055] [Example of synthesis 5] Other than using 104.0 kg 6-hydroxy capronic acid instead of lactic acid, similar method with synthesis example 1 was adopted and white powder form polycapronic acid was obtained. The amount obtained was 81.0 kg, yield was 92.2 % and weight average molecular weight (Mw) was 131,000.

[0056] [Example of synthesis 6] 8 kg polylactic acid obtained from the synthesis example 1 and 2 kg poly caprolacton were mixed well and a mixture of polylactic acid and polycaprolacton was obtained.

[0057] [Example of synthesis 7] Other than that reaction mass (75 kg polylactic acid, 225.0 kg diphenyl ether) of polylactic acid having 22,000 weight average molecular weight (Mw) obtained from similar method with synthesis example 1 was charged with 9.05 kg polycaprolacton having 131,000 weight average molecular weight which was obtained from synthesis example 4 and reacted furthermore for 29 hours at 130 °C/17mmHg, similar method was adopted with synthesis example 1 and block copolymer of polylactic acid and polycaprolacton (lactic acid content was 89 weight %) was obtained. The amount obtained was 78.7 kg, yield was 93.6 % and weight average molecular weight (Mw) was 151,000.

[0058] [Embodiment example 1] 7 kg polylactic acid obtained from the synthesis example 1 and 3 kg polyethylene glycol (molecular weight Mw=150,000) were mixed and 200 g inorganic additive SiO<sub>2</sub> (Aerozil 200, Nippon Aerozil Co., made) and 100 g aliphatic group carboxylic acid amide (erucic acid amide) was added and mixed well by Henschel Mixer. Obtained powder was extruded with biaxial extruder (36mm) at the temperature of 170°C and cut with Peletizer. After this pellet was dried for 10 hours at 60°C, it was formed by 40 mm inflation forming machine (dice diameter 40 mm) at the temperature of 160 to 170°C and inflation film having a folding

diameter of 150 mm and thickness of 30 to 40 μm was formed and rolled. This film did not cause blocking and opening property was excellent also. Elasticity was 7000kgf/cm<sup>2</sup> and anti blocking was excellent and slipping angle was 20°C.

[0059] [Embodiment example 2 to 11] Except changing aliphatic group polyester, polyalkylene glycol, inorganic additive and aliphatic group carboxylic amide to the value as shown in Table-1, they were done similarly with embodiment example 1. In either example, forming property was excellent and obtained film did not cause blocking and opening property was excellent. In Table 1-2, result of evaluation on forming property, anti blocking, slipping angle is shown.

[0060] [Comparative example 1 to 9] Except changing aliphatic group polyester, polyalkylene glycol, inorganic additive and aliphatic group carboxylic amide to the value as shown in Table-1, they were done similarly with embodiment example 1. In either case, forming property was bad by locating to the screw at the time of inflation forming and stability of bubble was not good and the obtained film caused blocking and opening property was not good. The result of evaluation on forming property, anti-blocking property and slipping angle is shown.

[0061]

[Table 1]

Embodiment example 1 to 5

Polymer composition	Embodiment example No.		1	2	3	4	5
	A aliphatic group polyester	synthesis example weight (%)	1 70	←	←	←	1 80
	B polyalkylene glycol	Kind	PEG	←	←	←	PPEG
		Molecular weight (10,000)	15	2	0.4	←	0.4
		Weight (5%)	30	20	15	20	20
Additive	C aliphatic group carbox- ylic acid amide	Kind	EA	BA	SNT	SA	QA
		Amount added (parts)	1	←	←	←	←
		D inorganic additive	Kind	SiO <sub>2</sub> A	←	SiO <sub>2</sub> B	←
		Amount added (parts)	5	←	3	←	←
Forming property			excellent	←	←	←	←
Property	Elasticity (kgf/cm <sup>2</sup> )		3000	5000	7000	5000	5500
	Blocking property		excellent	←	←	←	←
	Slipping angle (°C)		20	22	24	22	21

[Legend in Table 1 to 4]

[←] means [same to the left]

[PEG] polyethylene glycol

PPEG polypropylene-polyethylene glycol (melting point 62°C )

SiO<sub>2</sub> A Aerosil 200 (made by Japan Aerosil)SiO<sub>2</sub> B Sairicia 350 (made by Fuji Sirisia)

EA erucic acid amide (made by Nippon Seika)

SA stearic acid amide (made by Nippon Seika)

OA oleyl acid amide (made by Nippon Seika)

BA behenic acid amide (made by Nippon Seika)

SNT N-stearyl erucic acid amide (made by Nippon Seika)

PNT N-oleyl palmito amide (made by Nippon Seika)

Table 2 Embodiment example 5 to 10

Polymer composition	Embodiment example No.		5	6	7	8	9	10
	A aliphatic group polyester	synthesis example weight (%)	2 90	3 80	4 90	5 ←	6 80	7 90
		B polyalkylene glycol	Kind	PEG	←	←	←	←
	Molecular weight (10,000)		0.4	←	←	←	←	←
Additive	C aliphatic group carboxylic acid amide	Weight (5%)	10	20	10	←	20	10
		Kind	PNT	EA	←	←	←	←
	Amount added (parts	1	1	1	←	←	←	
	D inorganic additive	Kind	SiO <sub>2</sub>	SiO <sub>2</sub> B	←	←	←	←
Amount added (parts		5	3	3	←	←	←	
Forming property			excellent	←	←	←	←	←
Property	Elasticity (kgf/cm <sup>2</sup> )		2500	4000	2500	3000	4500	3000
	Blocking property		excellent	←	←	←	←	←
	Slipping angle (°C)		23	25	22	21	24	24

Table 3 Comparative example 1 to 5

Polymer composition	Embodiment example No.		1	2	3	4	5
	A aliphatic group polyester	synthesis example weight (%)	1	←	←	2	3
			85	←	←	90	80
			B polyalkylene glycol	Kind	PEG	←	←
Molecular weight (10,000)	0.4	←		←	←	←	
	Weight (5%)	15		←	←	10	20
Additive	C aliphatic group carboxylic acid amide	Kind	0	SNT	0	←	←
		Amount added (parts)	0	1	0	←	←
	D inorganic additive	Kind	SiO <sub>2</sub> B	0	←	←	←
		Amount added (parts)	3	0	←	←	←
Forming property			No good	Fair	No good	←	←
Property	Elasticity (kgf/cm <sup>2</sup> )		7000	←	←	6500	4000
	Blocking property		Fair	←	←	←	←
	Slipping angle (°C)		60	55	70<	←	←

Table 4 Comparative example 6 to 9

Polymer composition	Embodiment example No.		6	7	8	9
	A aliphatic group polyester	synthesis example weight (%)	4	5	6	7
			80	90	80	90
			B polyalkylene glycol	Kind	PEG	←
Molecular weight (10,000)	0.4	←		←	←	
Weight (5%)	20	10		20	10	
Additive	C aliphatic group carboxylic acid amide	Kind	0	←	←	←
		Amount added (parts)	0	←	←	←
	D inorganic additive	Kind	0	←	←	←
		Amount added (parts)	0	←	←	←
Forming property			No good	←	←	←
Property	Elasticity (kgf/cm <sup>2</sup> )		3000	3000	4500	3000
	Blocking property		No good	←	←	←
	Slipping angle (°C)		70<	←	←	←

[0065]

[Effect of the invention]

(1) Resin composition of this invention is excellent in forming property at the time of forming (for example, such as extrusion forming, inflation forming) comparing to the case with single component (A).

(2) Film and sheet of this invention are significantly superior in anti-blocking property and slipping property compared with the case of single component (A).

(3) The edge of the film and sheet of this invention are easily peeled when formed into roll shape or layered shape compared to the case of single component of (A).

(4) The bag made of the film and sheet of this invention has excellent opening property when pulling the edge when it is made into roll shape, compared to the case of single component (A).

(5) The film and sheet of this invention have the same or better forming workability compared to the case of single component (A).

(6) The film and sheet of this invention have biodegradable property under the natural environment compared to the case of single component (A).

*\*Translator's note*

*These words (not found in the dictionary) marked with \* seems to be type miss.*

Continued from the front page

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